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Magnetostrictive aftereffects in a dilute 50Fe-50Ni-C alloy

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MAGNETOSTRICTIVE AFTEREFFECTS IN A DILUTE 50Fe-50Ni-C ALLOY

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PREFACE

Crystallographically equivalent interstitial sites are energetically equivalent when the host crystal is free of any fields. This situation is shown in Fig. P1 from which it may be appreciated that the difference of the free energy of the "crystal" with an interstitial "I" located in "x" and "y" sites, respectively, is equal to zero. This energy equivalence of the various interstitial sites is destroyed when the host crystal is subjected to fields such as strain fields, magnetization fields or a combination of both as may be appreciated from Figs. P2 through P4. In this case, the interstitials will preferentially occupy certain sites. This preferential occupation will lead to anelasticity, magnetic disaccommodation and magnetostrictive aftereffects as will be described later.

If an originally unstrained crystal in which the interstitials populate crystallographically equivalent sites equally is subjected to a strain, the interstitials will redistribute among those sites. This redistribution will give rise to an additional strain known as anelastic strain. The anelastic strain will develop as the interstitials diffuse through the host crystal and will, therefore, lag behind the elastic strain. Hence, anelastic effects are often referred to as aftereffects. Anelastic behaviour in materials under the influence of stresses far

below that at which plastic flow occurs is well known[1,2]. A favorite example is the small elastic aftereffect due to the redistribution of a few tenths of a percent interstitials, such as carbon, in iron. The damping associated with the redistribution of interstitials in metals can be as large as twenty times the background value. This is the reason for the importance of anelasticity as a metallurgical tool.

The magnitude of the elastic aftereffect as well as of the damping can be related to the properties of interstitials through an analysis of the elastic free energy per unit volume of a crystal containing interstitials. This elastic free energy is given by

$$F_e = F_o + \frac{1}{2} C_{ijk\ell} e_{ij} e_{k\ell} + \sigma_{ij} e_{ij} c^{(m)} + \frac{RT}{2VC_o} c^{(m)} c^{(m)} . \quad (P1)$$

In Eq. (P1) F_o is a constant denoting the specific free energy of the unstrained crystal. The second term represents the elastic free energy of the crystal without interstitials; the quantities e_{ij} and $C_{ijk\ell}$ are the strains and elastic constants, respectively. Repeated indices imply summation. The third term represents the elastic energy due to the redistribution of interstitials under strain. The quantity σ_{ij} characterizes the stress field of the interstitials and the $c^{(m)}$'s designate the excess interstitial concentrations in the respective m-sites.

Finally, the fourth term represents the configurational entropy of the interstitials in the quadratic approximation.

For the two dimensional cubic model crystal in Fig. P2, F_e is given by[3]

$$F_e = F_o + \frac{1}{2} C_{11}(e_{xx}^2+e_{yy}^2) + C_{12}(e_{xx}e_{yy}) + \frac{1}{2} C_{44}e_{xy}^2$$

$$+ \sigma(e_{xx}c^{(x)} + e_{yy}c^{(y)} + \frac{RT}{2VC_o}(c^{(x)2} + c^{(y)2}). \qquad (P2)$$

The magnitude of the elastic aftereffect, Δe_{ii}, follows from Eq. (P2), and the five equilibrium conditions

$$\frac{\partial F}{\partial e_{ij}} = \frac{\partial F}{\partial c^{(m)}} = 0, \quad i,j,m = x,y.$$

If it is assumed that no volume change occurs, i.e., $e_{xx}+e_{yy} = 0$, and no defect reactions take place, $c^{(x)}+c^{(y)} = 0$, the result is

$$\Delta e_{ii} = 2\sigma^2 \frac{VC_o}{RT} . \qquad (P3)$$

It can be seen from Eq. (P3) that an investigation of the elastic aftereffect will not yield the sign of the interstitial stress field σ. Neither will a study of the response of the defective crystal to the periodic stresses and strain, commonly referred to as damping. The sign of σ must, therefore, be obtained from other evidence. In the

case of carbon in iron, for example, it is usually agreed that the positioning of the carbon in tetragonal Fe-C martensite suggests a positive sign of σ.

When the host crystal is magnetized uniformly, the energy equivalence of crystallographically equivalent sites is destroyed due to the reorientation of atomic magnetic dipoles as can be seen from Fig. P3 which for simplicity refers to $0^{o}K$. In principle, the dimensions of the model crystal will also change as the magnetic field is applied. This effect will be considered later. Redistribution of the interstitial dipoles will thus occur and give rise to a magnetic phenomenon analogous to the elastic aftereffect, which is known as magnetic aftereffect[4]. This aftereffect is the delayed change of the magnetic anisotropy after the crystal has been magnetized. The specific magnetic free energy for this situation is given by[5]

$$F_m = F_o + K_{ijk\ell}\alpha_i\alpha_j\alpha_k\alpha_\ell + H_{ij}\,\alpha_i\alpha_j c^{(m)} + \frac{RT}{VC_o}\, c^{(m)} c^{(m)} . \quad (P4)$$

The constant F_o is the free energy of the unstrained crystal. The second term represents the magnetic anisotropy energy of the interstitial free model crystal with α_i and $K_{ijk\ell}$ designating the directional cosines of the magnetization and anisotropy constants, respectively. The third term denotes again the contribution of the defect, here H_{ij} is the internal anisotropy field due to the presence of the defect. A procedure equivalent to the one described

for the elastic aftereffect yields the magnitude of the magnetic aftereffect $\Delta\alpha_i^2$, as

$$\Delta\alpha_i^2 = \frac{H^2 V C_o}{RT} . \qquad (P5)$$

Again, it can be seen form Eq. (P5) that an investigation of the magnetic aftereffect alone yields only the magnitude of the defect field but not its sign.

Finally, the case of an interstitial in a ferromagnetic material is presented. Here both the spontaneous magnetization and magnetostrictive fields need both be considered. The free energy of a ferromagnetic material containing interstitials is a combination of the two previously described cases as may be appreciated from Fig. P4. Both aftereffects previously described occur but, since M and e are coupled, a magnetostrictive aftereffect due to the redistribution of the interstitials can be observed. This aftereffect is the lag of the strain after the direction of the magnetization has been changed.

The analysis of the magnetostrictive aftereffect starts from an expression of the total free energy[6],

$$F = F_o + F_e + F_m + F_{me} + \text{entropy} \qquad (P6)$$

where F_{me} is magnetoelastic coupling energy,

$$F_{me} = B_{ijk\ell} e_{ij} \alpha_k \alpha_\ell$$

and $B_{ijk\ell}$ are the magnetostrictive constants[7]. For the cubic two dimensional ferromagnetic model crystal, the specific total free energy of the crystal is then given by combining Eqs. (P1), (P4), (P6), and (P7), and by omitting redundant terms,

$$F = F_o + \frac{1}{2} C_{11}(e_{xx}^2+e_{yy}^2) + C_{12}(e_{xx}e_{yy}) + \frac{1}{2} C_{44}(e_{xy}^2)$$

$$+ B_1[(\alpha_1^2 - \tfrac{1}{3})\, e_{xx}+(\alpha_2^2 - \tfrac{1}{3})e_{yy}] + B_2\alpha_1\alpha_2 e_{xy}$$

$$+ \sigma(e_{xx}c^{(x)} + e_{yy}c^{(y)}) + H[(\alpha_1^2 - \tfrac{1}{3})\, c^{(x)}+(\alpha_2^2 - \tfrac{1}{3})c^{(y)}]$$

$$+ \frac{RT}{2VC_o}(c^{(x)2} + c^{(y)2}). \qquad \text{(P8)}$$

The equilibrium strains and excess interstitial concentrations can be computed by minimizing the free energy given by Eq. (P8) with the constraints $e_{xx}+e_{yy} = c^{(x)} + c^{(y)} = 0$. Physically this means that no defect reactions which would give rise to a change of the total number of interstitials under consideration and a volume change are considered. Upon neglecting higher orders of σ, the magnitude of the magnetostrictive aftereffect $\Delta\lambda_i$ is obtained,

$$\Delta\lambda_i = \frac{\sigma H}{C_{11}-C_{12}} \frac{VC_o}{RT} . \qquad \text{(P9)}$$

Eq. (P9) shows that the magnetostrictive aftereffect yields the sign of the defect parameters σ or H if the sign of one of the two is known. Thus, for example, the carbon interstitial defect in bcc iron has been completely characterized through investigations of the elastic[8], magnetic and magnetostrictive aftereffects[9] and of the structure of Fe-C martensite. In particular, a negative sign for H was obtained from the magnetostrictive after-effect[9]. Eq. (P9) also affords an estimate of the order of magnitude of the magnetostrictive aftereffect. Using the example of carbon in bcc iron[9] again where $H \simeq 10^{6} Jm^{-3}$, $\frac{\sigma V C_o}{RT} = 10^{-1}$, $\lambda \simeq 10^{-6}$, $B \simeq 10^{6} Jm^{-3}$, $C_{11}-C_{12} \simeq 10^{11} Jm^{-3}$, it is seen that $\Delta\lambda \cong 10^{-6}$. The magnetostrictive aftereffect is thus not a large effect and careful experimentation is required for its study.

In the following, the investigation of the magneto-strictive aftereffect in one particular fcc Fe-Ni-C alloy due to the redistribution of carbon interstitial will be described in a style suitable for publication. The goal of the study was to obtain the sign of the defect magnetic field in this particular structure and alloy, hitherto un-known.

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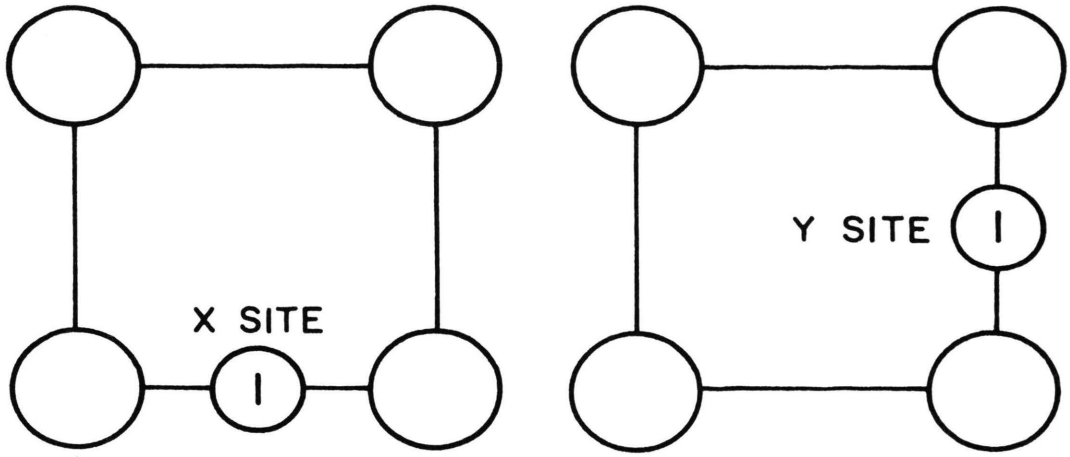
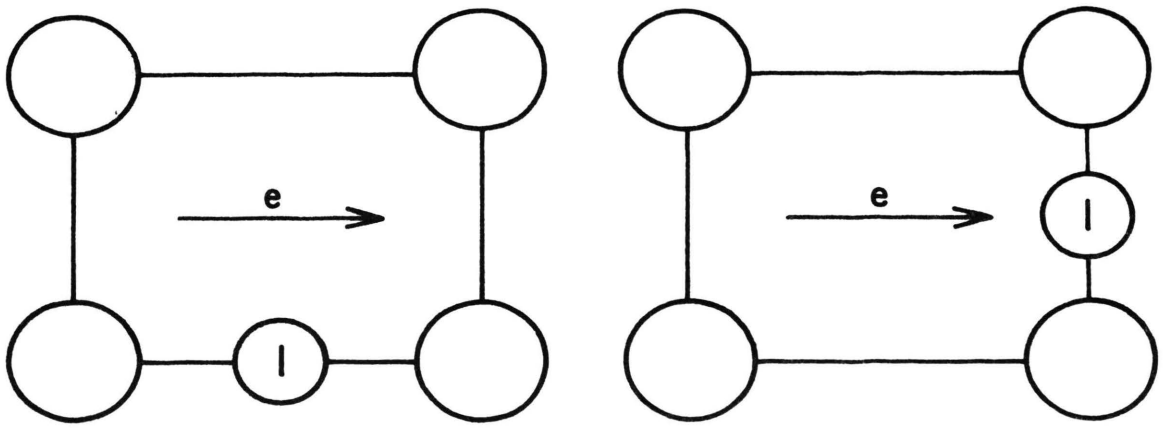


$\Delta F = 0$

Fig. P1. Free Energy Difference of a Two Dimensional Cubic Crystal With Interstitial "I" Located in "x" and "y" Sites.



$\Delta F = F_e$

Fig. P2. Free Energy Difference of a Two Dimensional Cubic Crystal With Interstitial "I", Under the Influence of Elastic Strains.

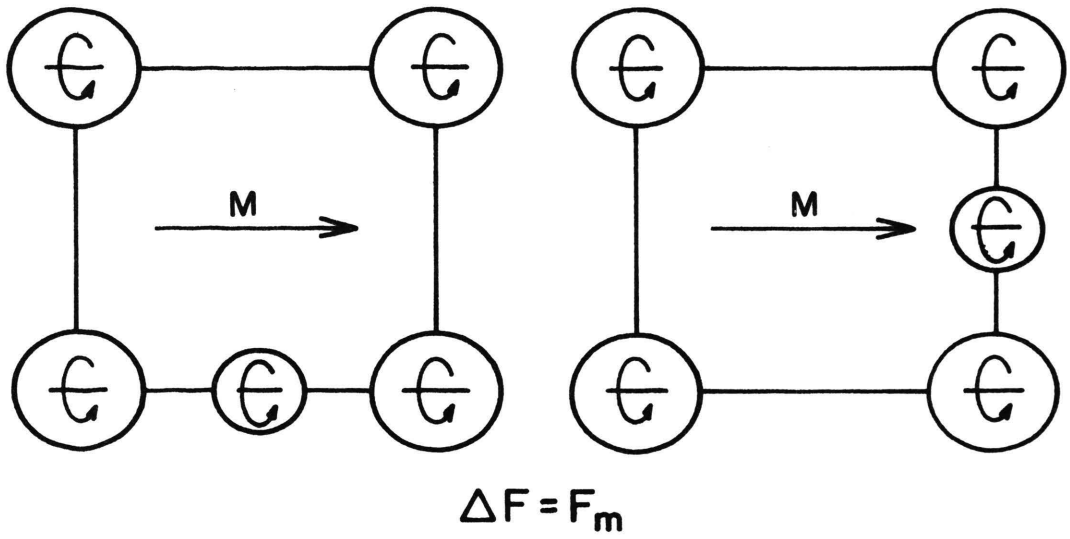


Fig. P3. Free Energy Difference of a Magnetized Two Dimensional Cubic Crystal With Interstitial "I" Under the Influence of Magnetic Strain.

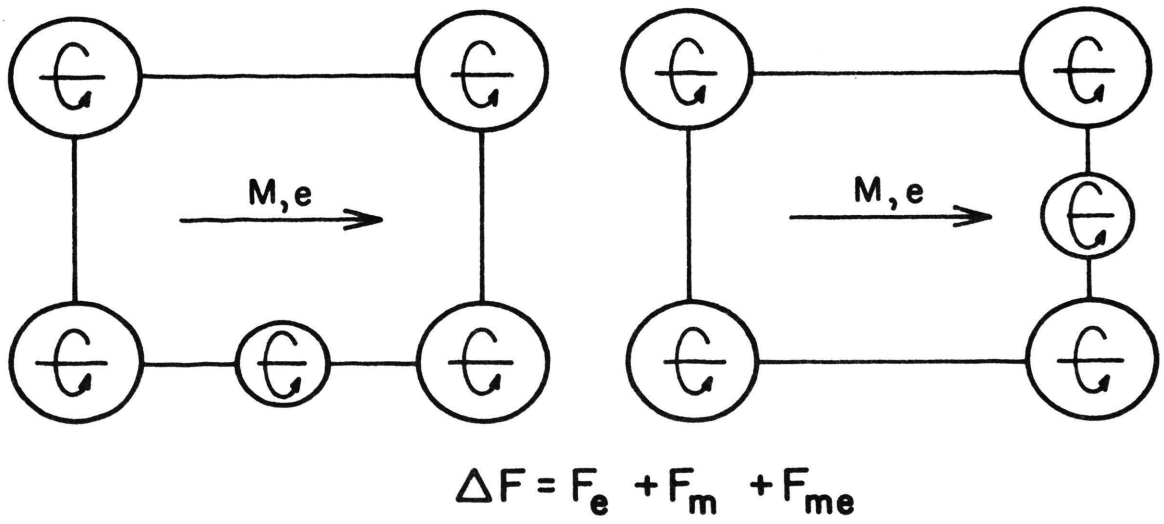


Fig. P4. Free Energy Difference of a Ferromagnetic Two Dimensional Cubic Crystal With Interstitial "I".

MAGNETOSTRICTIVE AFTEREFFECT IN A DILUTE 50Fe-50Ni-C ALLOY*

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INTRODUCTION

The specific free energy of a homogeneous strained ferromagnetic crystal at constant temperature containing a point defect is given in the harmonic approximation by[1,2]

$$F=F_o + \frac{1}{2} C_{ijk\ell}e_{ij}e_{k\ell} + B_{ijk\ell}e_{ij}\alpha_k\alpha_\ell + K_{ijk\ell}\alpha_i\alpha_j\alpha_k\alpha_\ell + \sigma_{ij}^{(m)}e_{ij}c^{(m)} + H_{ij}^{(m)}\alpha_i\alpha_j c^{(m)} + \frac{n_M RT}{2N} c^{(m)}c^{(m)} \qquad (1)$$

if defect reactions are neglected. In Eq. (1), standard notation has been used. The first four terms represent the free energy of the unstrained crystal and the strain, magnetoelastic and magnetocrystalline energies of the defect free crystal, respectively. The remaining terms

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*Submitted by Raj Master in partial fulfillment of the requirements for the M.S. degree in Metallurgical Engineering.

contain the contribution of the defects. The quantities $c^{(m)}$ denote the excess defect concentrations in the crystallographically equivalent m-sites of which n_M are energetically different. The defect property tensors σ and H represent the stress and anisotropy fields per defect. The magnitude of their components can be obtained from anelastic studies[3] which yield $|\sigma|$ or through investigations of the induced magnetic anisotropy[2] yielding $|H|$. A rough estimation of $|H|$ is also possible through stabilization field or disaccommodation measurements[4]. Neither of these studies, however, yields the sign of the defect property tensor components. The sign can be obtained from indirect evidence such as the structure of ordered defect phases from which the sign of σ_{ij} can be inferred or if the components of one property tensor are known, the sign of the components of the other property tensor can be obtained from a study of the magnetostrictive aftereffects[5]. Such a study has been performed on bcc Fe-C and Fe-N alloys and it was shown that the sign of H is negative and $|H|>|\sigma|$, both cases signifying that the interstitials prefer smaller sites[6]. It is the purpose of this paper to present the results of a study on magnetostrictive aftereffect in Fe-Ni-C alloys. It will be seen that the sign of the components of the property tensor H of the defect giving rise to aftereffects around $130^{o}C$ is positive.

EXPERIMENTAL DETAILS AND RESULTS

The investigation consisted of the preparation of pseudo single crystalline Fe-Ni-C alloys and the measurement of their dimensional changes upon changing the direction of the spontaneous magnetization in the temperature range around 130°C.

Disk shaped samples of Fe-Ni-C, 0.25 inches in diameter and 0.006 inches thick, were prepared from cold rolled 50Fe-50Ni stock (unannealed Isoperm). These samples were carburized by packing in graphite at about 200 microns of air and annealing at 1000°C sufficiently long to insure a homogeneous carbon distribution. The carbon content was found by weighing to be 0.23 wt% which agrees satisfactorily with the Fe-Ni-C equilibrium diagram[7]. During carburization, a pronounced (001)[100] recrystallization texture developed as evidenced by subsequent x-ray analysis. Before being used for aftereffect measurements, the samples were annealed for 15 hours at 215°C to obtain a specific point defect configuration as will be discussed below.

A differential strain gauge technique was applied to measure the magnetostrictive aftereffect directly[6]. In this technique four matched strain gauges are attached to the sample pairwise parallel and perpendicular to each other and in the desired "crystallographic directions". The gauges form a wheatstone bridge and are interconnected such that homogeneous sample deformations do not unbalance the

bridge. The temperature coefficient of the gauge was about the same as the coefficient of thermal expansion of the base Isoperm. The average sample temperature was controlled to within 0.1 degrees. All these provisions insured that the long time stability of the overall system measured in terms of strain was better than 10^{-8}.

A permanent magnet supplying a reasonably homogeneous field at the position of the sample (10^3 Acm^{-1} ±5%) was used to fix the direction of the spontaneous magnetization parallel to the plane of the disk. Since the anisotropy field of Isoperm[8] is about 5 Acm^{-1} and the demagnetization factor of the sample in the field direction is approximately 10^{-2}, the strength of the magnetic field was sufficient for the present purposes.

The actual experiment consisted of equilibrating the sample at a given temperature and direction of magnetization and the subsequent observation of the magnetostrictive aftereffect after the direction of magnetization had been changed. The result of a typical experiment carried out at 140°C is shown in Fig. 1. In this case, the gauge were mounted in [110] type directions. It can be seen that a sizable aftereffect is observed, and that the sample expanded in the direction of the magnetization. The data in Fig. 1 are not corrected for the form effect as it is negligibly small for the present samples.

A positive [100] aftereffect was also observed but its magnitude was much smaller than the [110] effect shown in

Fig. 1. No magnetostrictive aftereffect was found in the temperature range of interest in uncarburized but recrystallized Isoperm.

The kinetics of the aftereffect do not conform to simple exponentials. An analysis of the temperature dependence of the effective relaxation time yielded an activation energy of the process giving rise to the magnetostrictive aftereffect of 29 k cal $mole^{-1}$ confirming a previous observation[9].

DISCUSSION

The present study was guided in such a way that the same defect configuration responsible for the disaccommodation and stabilization in dilute Isoperm carbon alloys gave rise to the magnetostrictive aftereffect. This configuration has been identified to be a mixture of [100] and [111] tetragonal and [100] and [110] orthorhombic Fe-Ni-C point defects[9]. This defect mixture will give rise to magnetostrictive aftereffects in all crystallographic directions. The magnitude of the aftereffect in any specific direction will depend on the magnitude of the defect property tensors and the relative concentration of the various types of point defects. Both of these features are qualitatively inherent in a [110] type orthorhombic defect. Since this defect, shown in Fig. 2, also reflects the average iron-nickel concentration, this discussion will be couched in terms of its properties. Such a conceptual approximation is further justified by the fact that only certain general features of the defect were to be extracted from the data of the experiment described above.

The magnitude of the magnetostrictive aftereffect is obtained by minimizing the free energy given by Eq. (1) with respect to the strains and excess defect concentrations. The result for the effect in the [110] direction, $\Delta\lambda_{110}$, is

$$\Delta\lambda_{110} = \frac{1}{16RT}\left[\frac{3\sigma_1 H_1}{C_{11}-C_{12}} + \frac{8\sigma_2 H_2}{C_{44}}\right] \qquad (2)$$

if the difference between the unrelaxed and relaxed elastic stiffness constants is neglected. In Eq. (2) the quantities σ_1 and H_1 represent the defect "hydrostatic" planar stress and anisotropy fields of the [110] orthorhombic defect and σ_2 and H_2 denote the shear component of the defect fields.

From Eq. (2) it can be seen that the sign of the magnetostrictive aftereffect $\Delta\lambda_{110}$ can be both positive or negative depending on the signs and magnitudes of the two principle components of the two defect fields. The experimental results $\text{sgn}(\Delta\lambda_{110}) = \text{sgn}(\Delta\lambda_{100})$ and $|\Delta\lambda_{110}| > |\Delta\lambda_{100}|$, thus indicate that the signs of the products $\sigma_1 H_1$ and $\sigma_2 H_2$ are positive as well. Deductions about the signs of the compnents σ_1, σ_2, H_1 and H_2 are only possible if assumptions are made about the nature of the orthorhombic defect. For instance, it would appear reasonable to assume that $\sigma_1 < 0$ and/or $\sigma_2 < 0$ if a vacancy pair would replace either an iron or nickel pair of the defect[10]. This assumption would result in $H_1 < 0$ and/or $H_2 < 0$. On the other hand, little can be said if no vacancies are involved. Unpaired vacancies would lower the defect symmetry and render the situation more complex.

The magnitude of the magnetostrictive aftereffect $\Delta\lambda_{110}$ may be compared to the known magnetic anisotropy and internal friction in Fe-Ni-C alloys and their counterparts in Fe-C alloys. If details of the defect configuration are neglected for an order of magnitude estimation, it follows from Eq. (2) that

$$\Delta\lambda_{(Fe\text{-}Ni\text{-}C)} = \frac{\sigma(Fe\text{-}Ni\text{-}C)}{\sigma(Fe\text{-}C)} \cdot \frac{H(Fe\text{-}Ni\text{-}C)}{H(Fe\text{-}C)} \cdot \frac{C(Fe\text{-}Ni\text{-}C)}{C(Fe\text{-}C)} \Delta\lambda(Fe\text{-}C)$$

Using the values,[3,9,11,12]

$$\sigma(Fe\text{-}C) \simeq 10^{11}\ Jm^{-3}$$

$$\sigma(Fe\text{-}Ni\text{-}C) \simeq 0.3\sigma Fe\text{-}C$$

$$H(Fe\text{-}C) \simeq 6\times10^{6} Jm^{-3}$$

$$H(Fe\text{-}Ni\text{-}C) \simeq 2\times10^{6} Jm^{-3}$$

$$C(Fe\text{-}C) \simeq 0.008\ \omega t\%$$

$$C(Fe\text{-}Ni\text{-}C) \simeq 0.2\ \omega t\%$$

$$\Delta\lambda(Fe\text{-}C) \simeq 10^{-6}$$

it is found that

$$\Delta\lambda(Fe\text{-}Ni\text{-}C) \simeq 2\times10^{-6}$$

which agrees satisfactorily with the experimental value $\Delta\lambda(0)-\Delta\lambda(\infty) = 3.5\times10^{-6}$ taken from Fig. 1.

The main result of the present study is the positive sign of the magnetostrictive aftereffect in dilute fcc Isoperm carbon alloys, just the opposite from the bcc Fe-C,N cases. In the latter, the sign and magnitude of the magnetostrictive aftereffect could be used to determine directly the defect property tensor component equivalent to H_2 in

Eq. (2). This is not so in Fe-Ni-C alloys with their more complicated defect structure. For their complete characterization, measurements of the carbon induced anelasticity and magnetostrictive aftereffect in Fe-Ni single crystals of various Ni contents would have to be made to fully complement the anisotropy data[9].

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FIGURE CAPTIONS

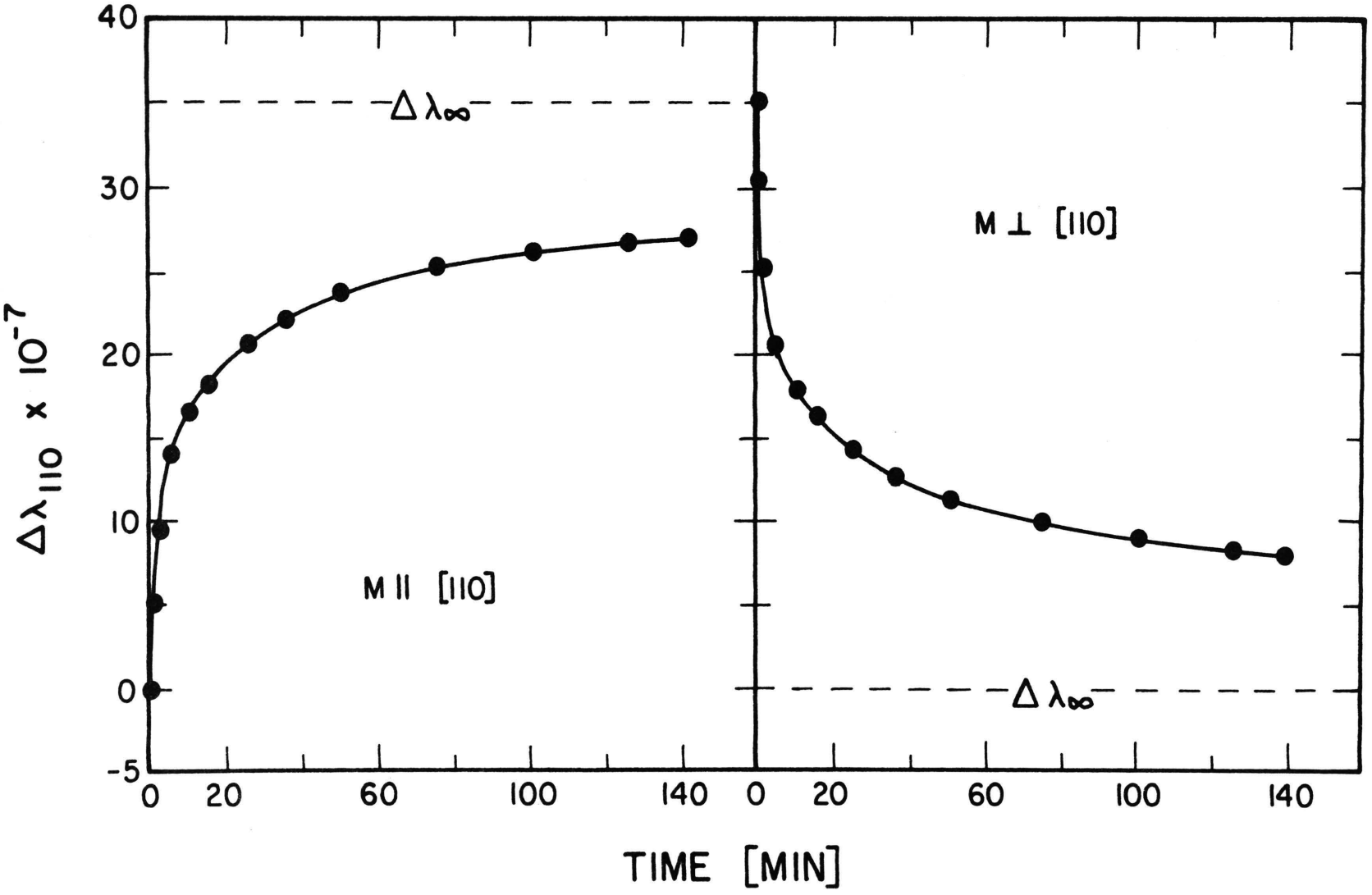

Fig. 1. Change in Magnetostrictive Constant $\Delta\lambda_{110}$ of a 50Fe-toNi-0.23C Alloy, As a Function of Time Measured at 140°C.

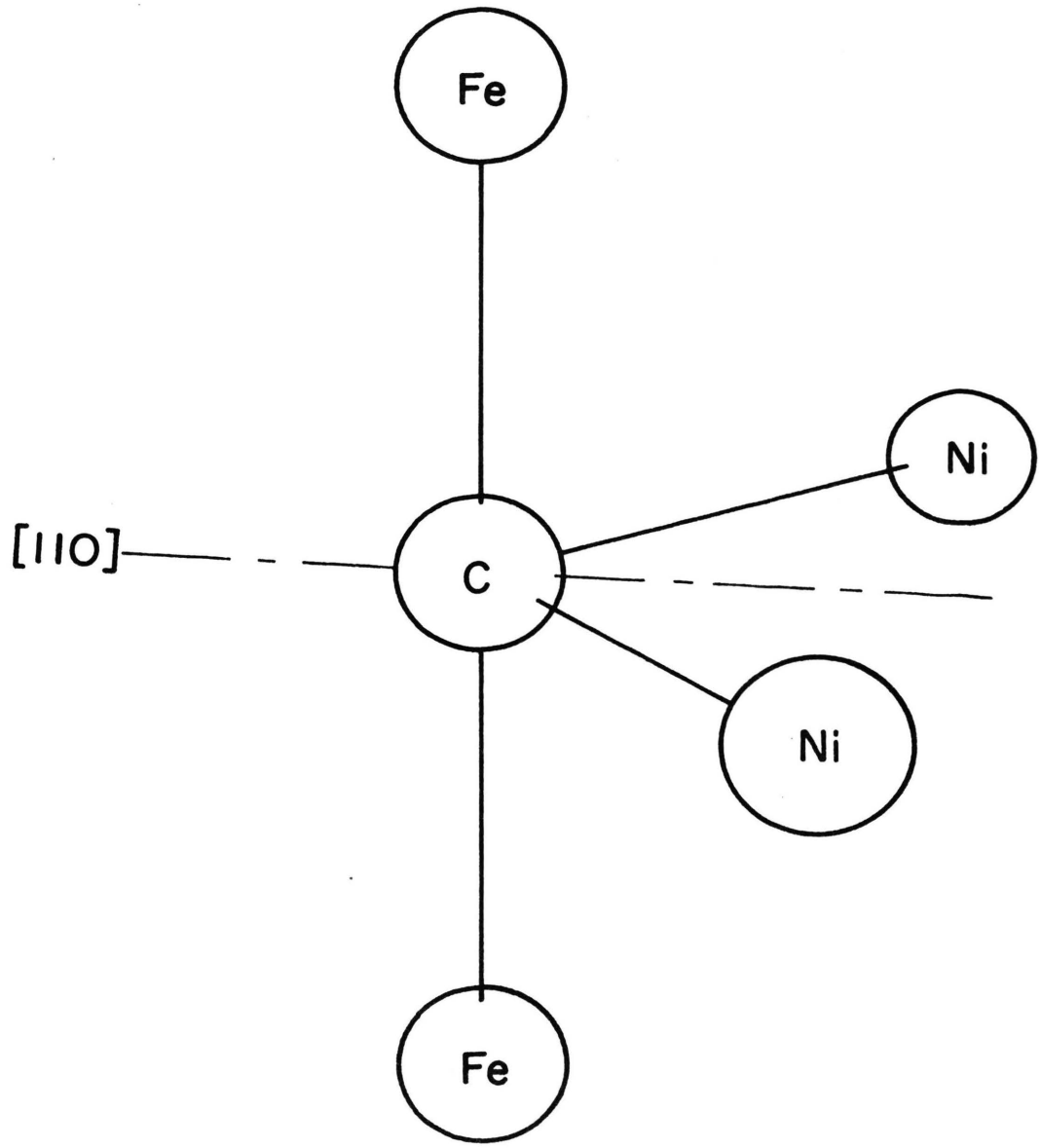

Fig. 2. A Typical [110] Orthohombic Defect Configuration.

VITA

Raj Navinchandra Master was born on March 8, 1950, in Bombay, India. He received his primary and secondary education in Bombay, India. He has received his college education from St. Xavier's College, Bombay, India; Faculty of Technology and Engineering, M.S., University, Baroda, India, from which he received a Bachelor of Engineering degree in Metallurgical Engineering in June 1972.

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